

D E C L A R A T I O N

I, Hideki OMOTE, a national of Japan, c/o Sumitomo Chemical Intellectual Property Service, Limited, 5-33, Kitahama 4-chome, Chuo-ku, Osaka-shi, Osaka 541-8550, Japan, declare that to the best of my knowledge and belief the attached is a full, true and faithful translation into English made by me of the certified copy of Japanese Patent Application No. Heisei 11-370676 attached thereto.

Signed this 22nd day of October, 2004


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[Title of the Invention] Catalyst for addition polymerization
and process for producing addition polymer

[Scope of Claims for Patent]

5 [Claim 1]

A catalyst for addition polymerization obtained by
contacting (A), (B) and (C) described below:

(A) an organoaluminum compound,

(B) one or more of boron compounds selected from (C1) to
10 (C3) below;

(B1) a boron compound represented by the general formula
 $BQ^1Q^2Q^3$,

(B2) a boron compound represented by the general formula
 $G^+(BQ^1Q^2Q^3Q^4)^-$, and

15 (B3) a boron compound represented by the general formula
 $(L-H)^+(BQ^1Q^2Q^3Q^4)^-$

(wherein B is a boron atom, Q^1 to Q^4 are a halogen atom, a
hydrocarbon group, a halogenated hydrocarbon group, a
heterocyclic group, a substituted silyl group, an alkoxy group,
20 or a di-substituted amino group, and they may be the same
or different. G^+ is an inorganic, organic or organometallic
cation. L is a neutral Lewis base, and $(L-H)^+$ is a Brønsted
acid.), and

(C) a solid inorganic compound.

25 [Claim 2]

The catalyst for addition polymerization according to
claim 1, wherein the organoaluminum compound is one or more
of aluminum compounds selected from (A1) to (A4) below:

(A1) An organoaluminum compound represented by the general formula; $R_rAl(OR)_oH_pX^1_q$;

(A2) an organoaluminum compound represented by the general formula; M^1AlR_4 ;

5 (A3) a cyclic aluminoxane having a structure represented by the general formula; $\{-Al(R)-O-\}_j$; and

(A4) an aluminoxane having a structure represented by the general formula; $R\{-Al(R)-O-\}_k AlR_2$

(wherein each of R's independently represents a hydrocarbon group having 1 to 20 carbon atoms; each of X^1 's independently represents a halogen atom; M^1 represents an alkaline metal atom; "r" represents a numeral satisfying $0 < r \leq 3$; "o" represents a numeral satisfying $0 \leq o < 3$; "p" represents a numeral satisfying $0 \leq p < 3$; "q" represents a numeral
10 satisfying $0 \leq q < 3$; $r + o + p + q = 3$; "j" represents an integer of 2 or more; and "k" represents an integer of 1 or more.)

[Claim 3]

The catalyst for addition polymerization according to
20 claim 1 or 2, wherein the solid inorganic compound (C) is a compound selected from the group consisting of metals of the Groups 1 to 3 and 6 to 16 of the Periodic Table of the Elements.

[Claim 4]

25 The catalyst for addition polymerization according to claim 1 or 2, wherein the solid inorganic compound is a Brønsted acid salt of a metal.

[Claim 5]

The catalyst for addition polymerization according to claim 1 or 2, wherein the solid inorganic compound is a halogenated metal compound.

[Claim 6]

5 A process for producing an addition polymer, which comprises using the catalyst for addition polymerization of any one of claims 1 to 5.

[Claim 7]

The process according to claim 6, wherein the addition
10 polymer is a copolymer of ethylene with an α -olefin.

[Detailed Description of the Invention]

[0001]

[Technical Field to Which the Invention Belongs]

The present invention relates to a catalyst for addition
15 polymerization and a process for producing an addition polymer.

[0002]

[Prior Art]

It is known that a polyethylene is obtained by contacting
20 ethylene with an alkylaluminum which has been used as a co-catalyst in "Angew. Chem. Vol.64, pp323(1952)" and "Makromol. Chem. Vol.193, pp1283(1992)".

Further, Reports relating to an addition polymerization using an aluminum complex as a catalyst have been known in
25 "J. Am. Chem. Soc. Vol.119, pp8125 (1997)", "J. Am. Chem. Soc. Vol.120, pp8277(1998)", "Chem. Commun., pp2523(1998)", and a specification of International Application WO98/40421.

A production process of an addition polymer with a catalyst

in which an organoaluminum compound is supported on a carrier, is also known. For example, JP-A-52-2890 as an example of being supported on a metal acetate, and "Polymer Preprints, Japan, Vol.46, pp216 (1997)" and "Polymer Preprints, Japan, 5 Vol.46, pp1215 (1997)" as an example of being supported on a metal chloride are known.

[0003]

[Problem to Be Solved by the Invention]

However, these catalysts for addition polymerization have 10 been not always satisfied from the viewpoint of catalyst activity.

In view of the above-situations, a subject of the present invention, namely, an object is to provide a catalyst for addition polymerization showing a highly polymerization 15 activity, and a process for producing efficiently an addition polymer.

[0004]

[Means for Solving the Problem]

Namely, the present invention relates to a catalyst for 20 addition polymerization obtained by contacting (A), (B) and (C) described below, and to a process for producing an addition polymer with the catalyst:

(A) an organoaluminum compound,

(B) one or more of boron compounds selected from (C1) to 25 (C3) below;

(B1) a boron compound represented by the general formula $BQ^1Q^2Q^3$,

(B2) a boron compound represented by the general formula

$G^+(BQ^1Q^2Q^3Q^4)^-$, and

(B3) a boron compound represented by the general formula $(L-H)^+(BQ^1Q^2Q^3Q^4)^-$.

(wherein B is a boron atom, Q^1 to Q^4 are a halogen atom, a hydrocarbon group, a halogenated hydrocarbon group, a heterocyclic group, a substituted silyl group, an alkoxy group, or a di-substituted amino group, and they may be the same or different. G^+ is an inorganic, organic or organometallic cation; L is a neutral Lewis base, and $(L-H)^+$ is a Brønsted acid.), and

(C) a solid inorganic compound.

The present invention is illustrated in detail below.

[0005]

[Mode for Practicing the Invention]

(A) Organoaluminum compound

The organoaluminum compound used in the present invention is an aluminum compound having an Al-C bond. Various such as aluminum compounds can be used, and one or more of aluminum compounds selected from (A1) to (A4) described below are preferable:

(A1) An organoaluminum compound represented by the general formula; $R_xAl(OR)_oH_pX_q^1$,

(A2) an organoaluminum compound represented by the general formula; M^1AlR_4 ,

(A3) a cyclic aluminoxane having a structure represented by the general formula; $\{-Al(R)-O-\}_j$, and

(A4) a linear aluminoxane having a structure represented by the general formula; $R\{-Al(R)-O-\}_k AlR_2$.

(wherein each of R's independently represents a hydrocarbon group having 1 to 20 carbon atoms; each of X¹'s independently represents a halogen atom; M¹ represents an alkaline metal atom; "r" represents a numeral satisfying $0 < r \leq 3$; "o" represents a numeral satisfying $0 \leq o < 3$; "p" represents a numeral satisfying $0 \leq p < 3$; "q" represents a numeral satisfying $0 \leq q < 3$; $r + o + p + q = 3$; "j" represents an integer of 2 or more; and "k" represents an integer of 1 or more.)

10 [0006]

As the organoaluminum compound (Al), the following compounds and the like can be exemplified.

① An organoaluminum compound represented by the general formula; $R_rAl(OR)_{3-r}$

15 (wherein each of R's independently represents a hydrocarbon group having 1 to 20 carbon atoms, preferably 1 to 15 carbon atoms, and more preferably 1 to 8 carbon atoms. "r" represents a numeral satisfying $0 < r \leq 3$, and preferably $1 \leq r \leq 3$.)

② An organoaluminum compound represented by the general formula; $R_rAlX^1_{3-r}$

(wherein each of R's independently represents a hydrocarbon group having 1 to 20 carbon atoms, preferably 1 to 15 carbon atoms, and more preferably 1 to 8 carbon atoms, and each of X¹'s independently represents a halogen atom. "r" represents a numeral satisfying $0 < r \leq 3$, and preferably $0 < r < 3$.)

③ An organoaluminum compound represented by the general formula; R_rAlH_{3-r}

(wherein each of R's independently represents a hydrocarbon

group having 1 to 20 carbon atoms, preferably 1 to 15 carbon atoms, and more preferably 1 to 8 carbon atoms. "r" represents a numeral satisfying $0 < r \leq 3$, and preferably $2 \leq r < 3$.)

④ An organoaluminum compound represented by the general
5 formula; $R_rAl(OR)_oX^1_q$,
(wherein each of R's independently represents a hydrocarbon group having 1 to 20 carbon atoms, preferably 1 to 15 carbon atoms, and more preferably 1 to 8 carbon atoms, and each of X^1 's independently represents a halogen atom. "r" represents
10 a numeral satisfying $0 < r \leq 3$, "o" represents a numeral satisfying $0 \leq o < 3$, "q" represents a numeral satisfying $0 \leq q < 3$, and $r + o + q = 3$.)

Each of R's in the general formula representing the above-mentioned organoaluminum compounds ① to ④ is
15 independently an alkyl group or an aryl group, and each of X^1 's is independently a chlorine atom or a bromine atom.

[0007]

Specific examples of the organoaluminum compound of the fore-mentioned ① include tri-n-alkylaluminums such as
20 trimethylaluminum, triethylaluminum, tri-n-butylaluminum, tri-n-hexylaluminum, tri-n-octylaluminum and the like; tri branched-alkyl aluminums such as triisopropylaluminum, triisobutylaluminum, tri-sec-butylaluminum, tri-tert-butylaluminum, tri-2-methylbutylaluminum,
25 tri-3-methylbutylaluminum, tri-3-methylpentylaluminum, tri-4-methylpentylaluminum, tri-2-methylhexylaluminum, tri-3-methylhexylaluminum, tri-2-ethylhexylaluminum and the like; tricycloalkylaluminums such as

tricyclohexylaluminum and the like; triarylaluminums such as triphenylaluminum, tritolylaluminum and the like; trialkenylaluminums such as triisoprenylaluminum and the like; alkylaluminum dialkoxides such as isobutylaluminum dimethoxide, isobutylaluminum diethoxide, isobutylaluminum diisopropoxide and the like; dialkylaluminum alkoxides such as diethylaluminum ethoxide, dibutylaluminum butoxide and the like; alkylaluminum sesquialkoxides such as ethylaluminum sesquibutoxide, butylaluminum sesquibutoxide and the like; alkylaluminums whose alkyls are partially replaced with aryloxy group such as methylaluminum bis(2,6-di-tert-butyl-4-methylphenoxide), ethylaluminum bis(2,6-di-tert-butyl-4-methylphenoxide) and the like; partially alkoxyated alkylaluminums which have a mean composition in which r in the general formula representing the organoaluminum compound of the fore-mentioned ① is 2.5; etc.

[0008]

Specific examples of the organoaluminum compound of the fore-mentioned ② include alkylaluminum sesquihalides such as ethylaluminum sesquichloride, butylaluminum sesquichloride, ethylaluminum sesquibromide and the like; dialkylaluminum halides such as dimethylaluminum chloride, diethylaluminum chloride, dipropylaluminum chloride, diisobutylaluminum chloride, dihexylaluminum chloride, butylaluminum bromide and the like; alkylaluminum dihalides such as methylaluminum dichloride, ethylaluminum

dichloride, propylaluminum dichloride, isobutylaluminum dichloride, hexylaluminum dichloride, octylaluminum dichloride, butylaluminum dibromide and the like; etc.

[0009]

5 Specific examples of the organoaluminum compound of the fore-mentioned ③ include dialkylaluminum hydrides such as diethylaluminum hydride, dibutylaluminum hydride, diisobutylaluminum hydride and the like; alkylaluminum dihydrides such as ethylaluminum dihydride, propylaluminum
10 dihydride and the like; alkylaluminums whose alkyl is partially substituted with hydrogen.

[0010]

Specific examples of the organoaluminum compound of the fore-mentioned ④ include alkylaluminums which are
15 partially alkoxyated and halogenated, etc.

[0011]

The fore-mentioned organoaluminum compound (A2) is an organoaluminum compound represented by the general formula; M^1AlR_4 . Herein, M^1 represents an alkaline metal atom. The
20 alkaline metal atom is preferably a lithium atom, a sodium atom or a potassium atom, and preferably a lithium atom in particular. The organoaluminum compound (A2) is preferably $LiAl(C_2H_5)_4$ or $LiAl(C_7H_{15})_4$ in particular.

[0012]

25 As specific examples of R in a cyclic aluminoxane (A3) having the general formula; $\{-Al(R)-O-\}_j$ and a linear aluminoxane (A4) having the general formula; $R\{-Al(R)-O-\}_k AlR_2$, can be exemplified alkyl groups such as a methyl group,

an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, an isobutyl group, a n-pentyl group, a neopentyl group and the like. "j" is an integer of 2 or more and "k" is an integer of 1 or more. R is preferably a methyl group or an isobutyl group, j is 2 to 40, and k is 1 to 40.

[0013]

The above-mentioned aluminoxanes are prepared by various methods. The procedure is not particularly restricted, and they may be prepared according to well-known methods. For example, they are prepared by contacting a solution of a trialkylaluminum (for example, trimethylaluminum and the like) in a suitable organic solvent (aromatic hydrocarbon; e.g. benzene, aliphatic hydrocarbon; e.g. hexane; and the like with water. Further, a method for preparing the aluminoxanes by contacting trialkylaluminum (e.g. trimethylaluminum) with a metal salt containing water of crystallization (e.g. cupric sulfate hydrate) can be exemplified. It is considered that the aluminoxanes thus obtained are usually a mixture of (A3) and (A4).

[0014]

The organoaluminum compound used in the present invention is preferably the above-mentioned organoaluminum compound (A1), further preferably the above-mentioned organoaluminum compound ①, preferably a trialkylaluminum in particular among them, and most preferably trimethylaluminum, triethylaluminum, triisobutylaluminum, tri-n-hexylaluminum or tri-n-octylaluminum.

[0015]

(B) Boron compound

As the boron compound (B) used in the present invention, at least one compound selected from the group consisting of boron compounds (B1) represented by the general formula $BQ^1Q^2Q^3$,
5 boron compounds (B2) represented by the general formula $G^+(BQ^1Q^2Q^3Q^4)^-$ and boron compounds (B3) represented by the general formula $(L-H)^+(BQ^1Q^2Q^3Q^4)^-$ can be used.

[0016]

In the boron compound (B1) represented by the general
10 formula $BQ^1Q^2Q^3$, B represents a boron atom in the trivalent state; Q^1 to Q^3 are respectively a halogen atom, a hydrocarbon group, a halogenated hydrocarbon group, a substituted silyl group, an alkoxy group or a di-substituted amino group and they may be the same or different. Each of Q^1 to Q^3 is preferably
15 a halogen atom, a hydrocarbon group having 1 to 20 carbon atoms, a halogenated hydrocarbon group having 1 to 20 carbon atoms, a substituted silyl group having 1 to 20 carbon atoms, an alkoxy group having 1 to 20 carbon atoms or a di-substituted amino group having 2 to 20 carbon atoms, and each of more
20 preferable Q^1 to Q^3 is a halogen atom, a hydrocarbon group having 1 to 20 carbon atoms or a halogenated hydrocarbon group having 1 to 20 carbon atoms. Each of the more preferable Q^1 to Q^3 is a fluorinated hydrocarbon group having 1 to 20 carbon atoms which contains at least one fluorine atom, and in
25 particular, each of Q^1 to Q^3 is preferably a fluorinated aryl group having 6 to 20 carbon atoms which contains at least one fluorine atom.

[0017]

Specific examples of the compound (B1) include
tris(pentafluorophenyl)borane,
tris(2,3,5,6-tetrafluorophenyl)borane,
tris(2,3,4,5-tetrafluorophenyl)borane,
5 tris(3,4,5-trifluorophenyl)borane,
tris(2,3,4-trifluorophenyl)borane,
phenylbis(pentafluorophenyl) borane and the like, and
tris(pentafluorophenyl)borane is most preferable.

[0018]

10 In the boron compound (B2) represented by the general
formula $G^+(BQ^1Q^2Q^3Q^4)^-$, G^+ is an inorganic or organic cation;
B is a boron atom in the trivalent -state; and Q^1 to Q^4 are
the same as defined in Q^1 to Q^3 in the above-mentioned (B1).

[0019]

15 Specific examples of G^+ as the inorganic cation in the
compound represented by the general formula $G^+(BQ^1Q^2Q^3Q^4)^-$
include inorganic cations such as a lithium cation, sodium
cation, potassium cation, silver cation and the like, and
organometallic cations such as a ferrocenium cation, an
20 alkyl-substituted ferrocenium cation and the like, and as
the organic cation, a tetraalkylphosphonium cation,
tetraarylphosphonium cation, tetraalkylammonium,
trialkylsulfonium cation, diaryliodonium cation,
trialkylcarbenium cation and the like are illustrated. G^+
25 is preferably a carbenium cation, and a triphenylcarbenium
cation is particularly preferred. As the $(BQ^1Q^2Q^3Q^4)^-$,
tetrakis(pentafluorophenyl)borate,
tetrakis(2,3,5,6-tetrafluorophenyl)borate,

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tetrakis(2,3,4,5-tetrafluorophenyl)borate,

tetrakis(3,4,5-trifluorophenyl)borate,

tetrakis(2,3,4-trifluorophenyl)borate,

phenyltris(pentafluorophenyl)borate,

- 5 tetrakis(3,5-bistrifluoromethylphenyl)borate and the like
are mentioned.

[0020]

These specific combinations include lithium

tetrakis(3,5-bistrifluoromethylphenyl)borate, sodium

- 10 tetrakis(3,5-bistrifluoromethylphenyl)borate, potassium

tetrakis(3,5-bistrifluoromethylphenyl)borate, silver

tetrakis(pentafluorophenyl)borate, ferrocenium

tetrakis(pentafluorophenyl)borate,

1,1'-dimethylferrocenium tetrakis(pentafluorophenyl)

- 15 borate, tetrabutylphosphonium

tetrakis(pentafluorophenyl)borate, tetraphenylphosphonium

tetrakis(pentafluorophenyl)borate, tetramethylammonium

tetrakis(pentafluorophenyl) borate, trimethylsulfonium

tetrakis(pentafluorophenyl) borate, diphenyliodonium

- 20 tetrakis(pentafluorophenyl) borate, triphenylcarbenium

tetrakis(pentafluorophenyl) borate, triphenylcarbenium

tetrakis(3,5-bistrifluoromethylphenyl)borate and the like,

and triphenylcarbeniumtetrakis(pentafluorophenyl) borate
is most preferable.

- 25 [0021]

Further, in the boron compound (B3) represented by the
formula $(L-H)^+(BQ^1Q^2Q^3Q^4)^-$, L is a neutral Lewis base; $(L-H)^+$
is a Brønsted acid; B is a boron atom in the trivalent state;

and Q^1 to Q^4 are the same as Q^1 to Q^3 in the above-mentioned Lewis acid (B1).

[0022]

Specific examples of $(L-H)^+$ as the Brønsted acid in the
 5 compound represented by the formula $(L-H)^+(BQ^1Q^2Q^3Q^4)^-$ include a trialkyl-substituted ammonium, an N,N-dialkylanilinium, a dialkylammonium, a triarylphosphonium and the like, and examples of $(BQ^1Q^2Q^3Q^4)^-$ include those as previously described.

[0023]

10 These specific combinations include triethylammonium tetrakis(pentafluorophenyl)borate, tripropylammonium tetrakis(pentafluorophenyl)borate, tri(n-butyl)ammonium tetrakis(pentafluorophenyl)borate, tri(n-butyl)ammonium tetrakis(3,5-bistrifluoromethylphenyl)borate,
 15 N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate, N,N-diethylanilinium tetrakis(pentafluorophenyl)borate, N,N-2,4,6-pentamethylanilinium tetrakis(pentafluorophenyl) borate, N,N-dimethylanilinium tetrakis(3,5-bistrifluoromethylphenyl)borate,
 20 diisopropylammonium tetrakis(pentafluorophenyl)borate, dicyclohexylammonium tetrakis(pentafluorophenyl)borate, triphenylphosphonium tetrakis(pentafluorophenyl)borate, tri(methylphenyl)phosphonium tetrakis(pentafluorophenyl) borate, tri(dimethylphenyl)phosphonium
 25 tetrakis(pentafluorophenyl)borate and the like, and tri(n-butyl)ammonium tetrakis(pentafluorophenyl)borate or N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate is most preferable.

[0024]

As the boron compound used in the present invention, the boron compound (B2) or (B3) is preferable, and triphenylcarbeniumtetrakis(pentafluorophenyl)borate, tri(n-butyl)ammonium tetrakis(pentafluorophenyl) borate or N,N-dimethylanilinum tetrakis (pentafluorophenyl)borate is most preferable.

[0025]

(C) Solid inorganic compound

When the solid inorganic compound is used in the present invention, the solid inorganic compound is an inorganic compound which is solid at normal temperature under normal pressure, and compounds of a metal selected from the Groups 1 to 3 and 6 to 16 of the Periodic Table of the Elements are preferable and it is more preferable to use a Brønsted acid salt.

[0026]

A metal used in the Brønsted acid salt is a metal selected from the Groups 1 to 3 and 6 to 16 of the Periodic Table of the Elements, and specific examples thereof include lithium, beryllium, sodium, magnesium, aluminum, potassium, calcium, scandium, chromium, manganese, iron, cobalt, nickel, copper, zinc, gallium, germanium, rubidium, strontium, yttrium, molybdenum, technetium, ruthenium, rhodium, palladium, silver, cadmium, indium, tin, antimony, cesium, barium, lanthanum, tungsten, rhenium, osmium, iridium, platinum, gold, mercury, thallium, lead, bismuth, polonium, francium, radium, actinium, cerium, praseodymium, neodymium, promethium,

samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, lutetium, thorium, protactinium, uranium, neptunium and the like, and sodium, magnesium, manganese, iron, cobalt or nickel is preferable.

5 [0027]

Further, the fore-mentioned Brønsted acid includes hydrofluoric acid, hydrochloric acid, hydrobromic acid, hydroiodic acid and the like.

[0028]

10 The metal halide compound is a halide of a metal atom represented by the general formula (1) described below:



(wherein M^2 represents a metal atom selected from the Groups 1 to 3 and 6 to 16 of the Periodic Table of the Elements and X^2 represents a halogen atom. "m" represents a numeral of 1 or more, and "n" represents a product of a valence number of the metal atom and "m".)

15

[0029]

M^2 represents a metal atom selected from the Groups 1 to 6 and 6 to 16 of the Periodic Table of the Elements, and includes a lithium atom, beryllium atom, sodium atom, magnesium atom, aluminum atom, potassium atom, calcium atom, scandium atom, chromium atom, manganese atom, iron atom, cobalt atom, nickel atom, copper atom, zinc atom, gallium atom, germanium atom, rubidium atom, strontium atom, yttrium atom, molybdenum atom, technetium atom, ruthenium atom, rhodium atom, palladium atom, silver atom, cadmium atom, indium atom, tin atom, antimony atom, cesium atom, barium atom, lanthanum atom, tungsten atom,

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rhenum atom, osmium atom, iridium atom, platinum atom, gold atom, mercury atom, thallium atom, lead atom, bismuth atom, polonium atom, francium atom, radium atom, actinium atom, cerium atom, praseodymium atom, neodymium atom, promethium atom, samarium, europium atom, gadolinium atom, terbium atom, dysprosium atom, holmium atom, erbium atom, thulium atom, ytterbium atom, lutetium atom, thorium atom, protactinium atom, uranium atom, neptunium atom and the like, and sodium atom, magnesium atom, manganese atom, iron atom, cobalt atom or nickel atom is preferable.

[0030]

X^2 in the above-mentioned general formula (1) represents a halogen atom, and specific examples thereof include a fluorine atom, chlorine atom, bromine atom, iodine atom or the like. A chlorine atom, bromine atom or iodine atom is preferable.

[0031]

Specific examples of the metal halide represented by the above-mentioned general formula (1) include metal fluorides such as zinc(II) fluoride, aluminum fluoride, antimony(III) fluoride, antimony(V) fluoride, yttrium fluoride, iridium(I) fluoride, iridium(II) fluoride, iridium(III) fluoride, iridium(IV) fluoride, iridium(V) fluoride, iridium(VI) fluoride, indium(III) fluoride, uranium(III) fluoride, uranium(IV) fluoride, uranium(V) fluoride, osmium(IV) fluoride, osmium(V) fluoride, osmium(VI) fluoride, cadmium fluoride, gadolinium(III) fluoride, potassium fluoride, gallium(III) fluoride, calcium fluoride, gold(III) fluoride,

silver(I) fluoride, silver(II) fluoride, chromium(II) fluoride, chromium(III) fluoride, chromium(IV) fluoride, chromium(V) fluoride, chromium(VI) fluoride, cobalt(II) fluoride, cobalt(III) fluoride, samarium(II) fluoride, samarium(III) fluoride, mercury(I) fluoride, mercury(II) fluoride, scandium fluoride, tin(II) fluoride, tin(IV) fluoride, strontium fluoride, cesium fluoride, cerium(III) fluoride, cerium (IV) fluoride, thallium(I) fluoride, thallium(III) fluoride, tungsten(IV) fluoride, tungsten(V) fluoride, tungsten(VI) fluoride, hexatantalum tetradecylfluoride, hexatantalum pentadecylfluoride, tantalum(III) fluoride, tantalum(IV) fluoride, tantalum(V) fluoride, thulium(III) fluoride, technetium(V) fluoride, technetium(VI) fluoride, iron(II) fluoride, iron(III) fluoride, diiron pentafluoride, copper(I) fluoride, copper(II) fluoride, thorium(IV) fluoride, sodium fluoride, lead(II) fluoride, lead(IV) fluoride, hexaniobium undecylfluoride, niobium(III) fluoride, niobium(IV) fluoride, niobium(V) fluoride, nickel(II) fluoride, neptunium(III) fluoride, neptunium(IV) fluoride, platinum(IV) fluoride, platinum(V) fluoride, platinum(VI) fluoride, palladium(II) fluoride, palladium(IV) fluoride, barium fluoride, bismuth(III) fluoride, beryllium fluoride, plutonium(III) fluoride, Beryllium fluoride, holmium(III) fluoride, magnesium fluoride, manganese(II) fluoride, manganese(III) fluoride, manganese(IV) fluoride, molybdenum(II) fluoride, molybdenum(III) fluoride, molybdenum(IV) fluoride, europium(II) fluoride,

europium(III) fluoride, lanthanum fluoride, lithium fluoride,
ruthenium(II) fluoride, ruthenium(III) fluoride, rubidium
fluoride, rhenium (IV), rhenium(V) fluoride, rhenium(VI)
fluoride, rhenium(VII) fluoride, rhodium(III) fluoride,
5 rhodium(IV) fluoride, rhodium(V) fluoride, rhodium(VI)
fluoride and the like;

[0032]

metal chlorides such as zinc(I) chloride, zinc(II)
chloride, aluminum chloride, antimony(III) chloride,
10 antimony(IV) chloride, yttrium(III) chloride, iridium(I)
chloride, iridium(II) chloride, iridium(III) chloride,
iridium(IV), indium(I) chloride, indium(II) chloride,
indium(III) chloride, uranium(III) chloride, uranium(IV)
chloride, uranium(V) chloride, uranium(VI) chloride,
15 osmium(III) chloride, osmium(IV) chloride, osmium(V)
chloride, cadmium(II) chloride, potassium chloride,
gallium(I) chloride, gallium(II) chloride, gallium(III)
chloride, calcium chloride, gold(I) chloride, gold(III)
chloride, gold dichloride, silver chloride, chromium(II)
20 chloride, chromium(III) chloride, chromium(IV) chloride,
cobalt(II) chloride, mercury(I) chloride, mercury(II)
chloride, scandium chloride, tin(II) chloride, tin(IV)
chloride, strontium chloride, cerium(III) chloride,
cerium(IV) chloride, thallium(I) chloride, thallium(III)
25 chloride, thallium dichloride, thallium trichloride,
tungsten(II) chloride, tungsten(III) chloride, tungsten(IV)
chloride, tungsten(V) chloride, tungsten(VI) chloride,
hexatantalum tetradecylchloride, hexatantalum

pentadecylchloride, tantalum(III) chloride, tantalum(IV) chloride, tantalum(V) chloride, technetium(IV) chloride, technetium(VI) chloride, iron(II) chloride, iron(III) chloride, copper(I) chloride, copper(II) chloride,
5 thorium(IV) chloride, sodium chloride, lead(II) chloride, lead(IV) chloride, hexaniobium undecylchloride, hexaniobium pentadecylchloride, triniobium octachloride, niobium(III) chloride, niobium(IV) chloride, niobium(V) chloride, nickel(II) chloride, neodymium(II) chloride, neodymium(III) chloride, neptunium(III) chloride, neptunium(IV) chloride,
10 platinum(II) chloride, platinum(IV) chloride, platinum trichloride, palladium(II) chloride, bismuth(III) chloride, praseodymium(III) chloride, plutonium(III) chloride, plutonium(IV) chloride, beryllium chloride, magnesium chloride, manganese(II) chloride, molybdenum(II) chloride,
15 molybdenum(III) chloride, molybdenum(IV) chloride, molybdenum chloride (V), europium(II) chloride, europium(III) chloride, lanthanum chloride, lithium chloride, ruthenium(III) chloride, rubidium chloride, rhenium(III) chloride, rhenium(IV) chloride, rhenium(V) chloride,
20 rhenium(VI) chloride, rhodium(I) chloride, rhodium(II) chloride, rhodium(III) chloride and the like;

[0033]

metal bromides such as zinc(II) bromide, aluminum bromide,
25 antimony bromide, yttrium bromide, iridium(I) bromide, iridium(II) bromide, iridium(III) bromide, iridium(IV) bromide, indium(I) bromide, indium dichloride, indium(III) bromide, uranium(III) bromide, uranium(IV) bromide,

uranium(V) bromide, osmium(III) bromide, osmium(IV) bromide,
 cadmium(II) bromide, potassium bromide, gallium(I) bromide,
 gallium dibromide, gallium(III) bromide, calcium bromide,
 gold(I) bromide, gold(III) bromide, silver bromide,
 5 chromium(II) bromide, chromium(III) bromide, mercury(I)
 bromide, mercury(II) bromide, scandium bromide, tin(II)
 bromide, tin(IV) bromide, strontium bromide, cesium bromide,
 cerium(III) bromide, thallium(I) bromide, thallium(III)
 bromide, tungsten(II) bromide, tungsten(III) bromide,
 10 tungsten(IV) bromide, tungsten(V) bromide, tungsten(VI)
 bromide, hexatantalum tetradecylbromide, hexatantalum
 pentadecylbromide, tantalum(III) bromide, tantalum(IV)
 bromide, tantalum(V) bromide, iron(II) bromide, iron(III)
 bromide, triiron octabromide, copper(I) bromide, copper(II)
 15 bromide, thorium(IV) bromide, sodium bromide, lead(II)
 bromide, hexaniobium tetradecylbromide, triniobium
 octabromide, niobium(III) bromide, niobium(IV) bromide,
 niobium(V) bromide, nickel(II) bromide, neptunium(III)
 bromide, neptunium(IV) bromide, platinum(II) bromide,
 20 platinum(IV) bromide, platinum tribromide, palladium(II)
 bromide, barium bromide, bismuth bromide, plutonium(III)
 bromide, beryllium bromide, magnesium bromide, manganese(II)
 bromide, molybdenum(II) bromide, molybdenum(III) bromide,
 molybdenum(IV) bromide, europium(II) bromide, europium(III)
 25 bromide, lanthanum bromide, lithium bromide, ruthenium(II)
 bromide, ruthenium(III) bromide, rubidium bromide,
 rhenium(III) bromide, rhenium(IV) bromide, rhenium(V)
 bromide, rhodium(II) bromide, rhodium(III) bromide and the

like;

[0034]

metal iodides such as zinc(II) iodide, aluminum iodide, diantimony tetraiodide, antimony(III) iodide, yttrium iodide,
5 iridium(I) iodide, iridium(II) iodide, iridium(III) iodide, iridium(IV) iodide, indium(I) iodide, indium(III) iodide, uranium(II) iodide, uranium(IV) iodide, osmium(I) iodide, osmium(II) iodide, osmium(III) iodide, cadmium iodide, potassium iodide, gallium(I) iodide, gallium(III) iodide,
10 calcium iodide, gold(I) iodide, gold(III) iodide, silver iodide, titanium(III) iodide, chromium(II) iodide, chromium(III) iodide, cobalt(I) iodide, cobalt(III) iodide, mercury(I) iodide, mercury(II) iodide, scandium iodide, tin(II) iodide, tin(IV) iodide, strontium iodide, cesium
15 iodide, cerium(II) iodide, cerium(III) iodide, thallium(I) iodide, thallium(III) iodide, tungsten(II) iodide, tungsten(III) iodide, tungsten(IV) iodide, tungsten(V) iodide, tungsten(VI) iodide, iron(II) iodide, iron(III) iodide, triiron octaiodide, copper(I) iodide, copper(II)
20 iodide, thorium(IV) iodide, sodium iodide, lead(II) iodide, hexaniobium tetradecyliodide, triniobium octaiodide, niobium(III) iodide, niobium(IV) iodide, niobium(V) iodide, nickel(II) iodide, neptunium(III) iodide, neptunium(IV) iodide, platinum(II) iodide, platinum(IV) iodide, platinum
25 triiodide, palladium(II) iodide, barium iodide, bismuth(III) iodide, plutonium(III) iodide, beryllium iodide, magnesium iodide, manganese(II) iodide, molybdenum(II) iodide, molybdenum(III) iodide, molybdenum(IV) iodide, europium(II)

iodide, europium(III) iodide, lanthanum iodide, lithium iodide, ruthenium(II) iodide, ruthenium(III) iodide, rubidium iodide, rhenium(III) iodide, rhenium(IV) iodide, rhenium(V) iodide, rhodium(II) iodide, rhodium(III) iodide
5 and the like.

The metal halide is preferably sodium chloride, sodium iodide, magnesium chloride, manganese(II) chloride, iron(II) chloride or nickel(II) bromide.

[0035]

10 When the catalyst for polymerization of the present invention is used, the amount of the organoaluminum compound (A) used can be selected from a wide range in which the concentration of the organoaluminum compound (A) in the polymerization system is usually 10^{-6} to 10 mol/l. A range
15 of from 10^{-5} to 1 mol/l is preferable.

[0036]

The amount of the boron compound (B) is used at an amount in which a molar ratio of the organoaluminum compound (A) to the boron compound (B) [(A)/(B)] is usually from 0.1 to
20 10000 and preferably from 0.5 to 5000.

[0037]

The amount of the solid inorganic compound (C) used can be selected at a wide range in which the concentration of the solid inorganic compound (C) in the polymerization system
25 is usually 0.001mg/l to 1000g/l. The preferable range is from 0.01mg/l to 100g/l.

[0038]

As the method of supplying the respective components in

a polymerization vessel, for example, they are supplied in an inert gas such as nitrogen, argon or the like under a moisture free condition. The catalyst components (A), (B) and (C) may be separately fed, or may be fed after previously
5 contacting.

[0039]

The polymerization is usually carried out at a temperature of from -80°C to 300°C , preferably from -40°C to 280°C , and more preferably from 20°C to 250°C .

10 The polymerization pressure is not particularly limited, and the pressure range is preferably from normal pressure to about 150 atm. from industrial and economical viewpoints. In general, the polymerization time is appropriately determined depending on kinds of a desired polymer and a
15 reaction apparatus, and the range is usually from 1 minute to 40 hours.

[0040]

In the polymerization process, Either of a continuous process and a batch-wise process are possible. Further, a
20 slurry polymerization or a solvent polymerization in which a hydrocarbon solvent such as propane, pentane, hexane, heptane, octane, toluene or xylene, is used, a liquid phase polymerization without using a solvent, or a gas phase polymerization can be conducted.

25 [0041]

The process for producing an addition polymer of the present invention is a process for producing an addition polymer using the above-mentioned catalyst for addition polymerization,

preferably a process for producing an addition polymer in which an olefin is homo-polymerized, or an olefin with another olefin and /or other polymerizable unsaturated compound are copolymerized.

5 [0042]

As olefins which can be applied to polymerization in the present invention, olefins having 2 to 20 carbon atoms, in particular, ethylene, α -olefins having 3 to 20 carbon atoms, diolefins having 4 to 20 carbon atoms and the like can be
10 used, and 2 or more of monomers can be also used at the same time. Specific examples of the olefins include straight chain olefins such as ethylene, propylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene and the like, branched olefins such as 3-methyl-1-butene,
15 3-methyl-1-pentene, 4-methyl-1-pentene and the like, vinylcyclohexene and the like, and the present invention should not be limited to the above-mentioned compounds.

[0043]

As other polymerizable unsaturated compounds, acrylic
20 acid ester compounds, methacrylic acid ester compounds and alkenyl aromatic hydrocarbons are listed.

[0044]

Specific examples of the acrylic acid ester compounds include methyl acrylate and compounds in which the methyl
25 group thereof is replaced with ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl or the like, and specific examples of the methacrylic acid ester compounds include methyl methacrylate and compounds in which the methyl group

thereof is replaced with ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl or the like.

[0045]

As the alkenyl aromatic hydrocarbon compounds, an alkenyl
5 compound having an aromatic hydrocarbon group having 6 to
25 carbon atoms is preferable. Specific examples of the
aromatic hydrocarbon group having 6 to 25 carbon atoms include
a phenyl group, tolyl group, xylyl group, tert-butylphenyl
group, vinylphenyl group, naphthyl group, phenanthryl group,
10 anthracenyl group and the like. A phenyl group, tolyl group,
xylyl group, tert-butylphenyl group, vinyl phenyl group and
naphthyl group are preferable.

Specific examples of the alkenyl aromatic hydrocarbon
include alkenylbenzenes such as styrene, 2-phenylpropylene,
15 2-phenylbutene and the like; alkylstyrenes such as
p-methylstyrene, m-methylstyrene, o-methylstyrene,
p-ethylstyrene, m-ethylstyrene, o-ethylstyrene,
2,4-dimethylstyrene, 2,5-dimethylstyrene,
3,4-dimethylstyrene, 3,5-dimethylstyrene,
20 3-methyl-5-ethylstyrene, p-tert-butylstyrene,
p-sec-butylstyrene and the like; vinyl naphthalenes such as
1-vinylnaphthalene and the like, etc.

[0046]

The catalyst for addition polymerization of the present
25 invention is suitably used as a catalyst for olefin
polymerization, and the production process of addition
polymers is suitable for production of olefin polymers. As
copolymers produced by the production process of the present

invention, preferably, a copolymer of ethylene with an α -olefin (among them, a linear low density polyethylene (LLDPE) is preferable) in particular, and specific examples of the combination of monomers constituting the copolymer include
5 ethylene and propylene, ethylene and 1-butene, ethylene and 1-hexene, ethylene and 1-octene, and the like, but the present invention should not be limited to these combinations.

[0047]

Further, in order to control the molecular weight of the
10 addition polymer obtained, a chain transfer agent such as hydrogen, a silane compound or the like can be also added.

[0048]

[EXAMPLE]

The present invention is further illustrated in detail
15 according to Examples below, but the present invention is not limited thereto.

Further, the measurement values of respective items in Examples were measured according to methods described below.

[0049]

20 (1) Intrinsic viscosity ($[\eta]$: dl/g)

It was measured at 135°C in a tetralin solution using an Ubbellohde viscometer.

[0050]

(2) Weight average molecular weight (M_w), number average
25 molecular weight (M_n), and molecular weight distribution (M_w/M_n):

They were measured under the following conditions according to a gel permeation chromatography (GPC). Further,

calibration curve was prepared using standard polystyrene. Molecular weight distribution was evaluated by a ratio (M_w/M_n) of the weight average molecular weight (M_w) to the number average molecular weight (M_n).

5 Machine: 150C type, manufactured by Milipore Waters Co., Ltd.

Column: Shodex M/S 80

Measurement temperature: 145°C

Solvent: O-dichlorobenzene

10 Sample concentration: 5 mg/8 ml

[0051]

(3) Melting point (T_m : °C)

It was measured using DSC-VII manufactured by Perkin Elmer Co., Ltd. according to the conditions below.

15 Heating: 40°C to 150°C (10°C/min.),
retaining for 5 minutes.

Cooling: 150°C to 40°C (5°C/min.),
retaining for 10 minutes.

Measurement: 40°C to 160°C (raised at 5°C /min.)

20 [0052]

(4) Comonomer unit composition in copolymer

¹³C-NMR was measured according to the conditions below using AM400 manufactured by Bruker Co., Ltd., and the comonomer unit composition in copolymer was determined by
25 calculating method which is described in J.C.Randall, J. Macro. Sci. -Rev. Macromol. Chem. Phys., Vol.C29(2&3), page 201, (1989).

Solvent: O-dichlorobenzene/deuterated benzene =4/1

-29-

(volume ratio)

Concentration: 200 mg/3 ml

Temperature: 135°C

[0053]

5 The respective compounds used at polymerization in
Examples described below are below.

· Triisobutylaluminum: commercially available product
manufactured by Tosoh-Akzo Co., Ltd. Triisobutylaluminum
was diluted with toluene to be used as a 1M solution.

10 · Triphenylcarbenium tetrakis(pentafluorophenyl) borate:
commercially available product manufactured by Tosoh-Akzo
Co., Ltd. Triphenylcarbenium tetrakis
(pentafluorophenyl) borate was diluted with toluene to be
used as a 0.005M solution.

15 · Manganese(II) chloride (manufactured by Aldrich Co., Ltd.,
purity=99.999%), iron(II) chloride (manufactured by Kanto
Chemicals Co., Ltd., extra pure grade), magnesium chloride
(manufactured by Junsei Chemicals Co., Ltd., purity=99.99%),
nickel(II) bromide (manufactured by Strem Chemicals Co., Ltd.,
20 purity=99%), sodium chloride (manufactured by Kanto Chemicals
Co., Ltd., extra pure grade), sodium iodide (manufactured by
Aldrich Co., Ltd., purity=99.999%) were milled under nitrogen
atmosphere using an agate mortar.

[0054]

25 [Example 1]

The atmosphere of a 400ml stirring type autoclave made
of stainless had been replaced with argon, 200ml of purified
toluene was charged thereto, and the inside of the autoclave

was adjusted at 35°C. Thereto, 1ml (1mmol) of triisobutylaluminum and 14mg of manganese(II) chloride were added. After stirring at 35°C for 3 minutes, 1ml (5 μ mol) of triphenylcarbenium tetrakis(pentafluorophenyl) borate was added and the mixture was stirred. The molar ratio [Al]/[B] of aluminum atom to boron atom was 200. Ethylene gas of 1.0 MPa was fed and polymerization was carried out at 40°C for 60 minutes. During the polymerization, ethylene gas was continuously fed so that the pressure in the system is constant. Then, the polymerization was terminated by introducing 10ml of ethanol under pressure. The unreacted ethylene gas was purged, the content of the autoclave was charged in 400ml of ethanol into which 20ml of 3N hydrochloric acid was added, a polymer precipitated was separated by filtration, and drying was carried out at 80°C for about 4 hours. As a result, 7.0g of a polyethylene was obtained. The intrinsic viscosity ($[\eta]$) of the polyethylene obtained was 1.39dl/g, weight average molecular weight (Mw) was 75600, and molecular weight distribution (Mw/Mn) was 2.0.

[0055]

[Example 2]

Polymerization was carried out in the same manner as in Example 1, except that manganese(II) chloride was changed to 9.5mg of iron(II) chloride. As a result, 2.07g of a polyethylene was obtained. The weight average molecular weight (Mw) of the polyethylene obtained was 75400, and molecular weight distribution (Mw/Mn) was 1.6.

[0056]

[Example 3]

Polymerization was carried out in the same manner as in Example 1, except that manganese(II) chloride was changed to 11.7mg of magnesium chloride. As a result, 5.24g of a polyethylene was obtained. The weight average molecular weight (Mw) of the polyethylene obtained was 75800, and molecular weight distribution (Mw/Mn) was 1.7.

[0057]

[Example 4]

Polymerization was carried out in the same manner as in Example 1, except that manganese(II) chloride was changed to 16.1mg of nickel(II) bromide. As a result, 1.75g of a polyethylene was obtained. The weight average molecular weight (Mw) of the polyethylene obtained was 78500, and molecular weight distribution (Mw/Mn) was 1.6.

[0058]

[Example 5]

Polymerization was carried out in the same manner as in Example 1, except that manganese(II) chloride was changed to 11.8mg of sodium chloride. As a result, 0.48g of a polyethylene was obtained. The weight average molecular weight (Mw) of the polyethylene obtained was 134000, and molecular weight distribution (Mw/Mn) was 2.6.

[0059]

[Example 6]

Polymerization was carried out in the same manner as in Example 1, except that manganese(II) chloride was changed to 13.5mg of sodium iodide. As a result, 0.63g of a polyethylene

was obtained. The weight average molecular weight (M_w) of the polyethylene obtained was 74300, and molecular weight distribution (M_w/M_n) was 1.6.

[0060]

5 [Example 7]

The atmosphere of a 400ml stirring type autoclave made of stainless had been replaced with argon, 100ml of purified toluene, 100ml of 1-hexene and ethylene gas of 1.0 MPa were charged thereto, and the inside of the autoclave was adjusted
10 at 40°C. Thereto, 1ml (1mmol) of triisobutylaluminum and 27.5mg of manganese(II) chloride were added. After stirring at 40°C for 3 minutes, 3ml (15 μ mol) of triphenylcarbenium tetrakis(pentafluorophenyl)borate was added and the mixture was stirred. The molar ratio $[Al]/[B]$ of aluminum atom to
15 boron atom was 67. Ethylene gas of 1.0 MPa was fed and polymerization was carried out at 40°C for 60 minutes. During the polymerization, ethylene gas was continuously fed so that the pressure in the system is constant. Then, the polymerization was terminated by introducing 10ml of ethanol
20 under pressure. The unreacted ethylene gas was purged, the content of the autoclave was charged in 400ml of ethanol into which 20ml of 3N hydrochloric acid was added, a polymer precipitated was separated by filtration, and drying was carried out at 80°C for about 4 hours. As a result, 1.36g of
25 a copolymer was obtained. The intrinsic viscosity ($[\eta]$) of the copolymer obtained was 1.15dl/g, weight average molecular weight (M_w) was 57400, and molecular weight distribution (M_w/M_n) was 1.7. The 1-hexene unit composition in the copolymer

was 0.11.

[0061]

[Comparative Example 1]

The atmosphere of a 400ml stirring type autoclave made
5 of stainless had been replaced with argon, 200ml of purified
toluene was charged thereto, and the inside of the autoclave
was adjusted at 35°C. Thereto, 1ml (1mmol) of
triisobutylaluminum and 13mg of manganese(II) chloride were
added. Ethylene gas of 1.0 MPa was fed and the system was
10 maintained at 40°C for 60 minutes. Then, 10ml of ethanol was
charged under pressure. The unreacted ethylene gas was purged,
and the content of the autoclave was charged in 400ml of ethanol
into which 20ml of 3N hydrochloric acid was added. As a result,
no polyethylene was obtained.

15 [0062]

[Comparative Example 2]

The atmosphere of a 400ml stirring type autoclave made
of stainless had been replaced with argon, 200ml of purified
toluene was charged thereto, and the inside of the autoclave
20 was adjusted at 35°C. Thereto, 10mg of manganese(II) chloride
and 1ml (5 μ mol) of triphenylcarbenium
tetrakis(pentafluorophenyl)borate were added. Ethylene gas
of 1.0 MPa was fed and the system was maintained at 40°C for
60 minutes. Then, 10ml of ethanol was charged under pressure.
25 The unreacted ethylene gas was purged, and the content of
the autoclave was charged in 400ml of ethanol into which 20ml
of 3N hydrochloric acid was added. As a result, no polyethylene
was obtained.

[0060]

[Effect of the Invention]

As described above in detail, according to the present invention, a catalyst for addition polymerization which
5 reveals a high polymerization activity, and a process for producing efficiently an addition polymer are provided. A high polymerization activity can be revealed according to the present invention without using a conventional solid catalyst component (a solid catalyst component in which a
10 titanium atom, a halogen atom and an electron donor are essential components), a transition metal compound such as a metallocene complex or the like which has been considered as essential for revealing a high polymerization activity, and an olefin polymer of high molecular weight can be produced,
15 therefore the value of the present invention is extremely great.

[Document Name] ABSTRACT

[Abstract]

[Subject] To provide a catalyst for addition polymerization showing a high polymerization activity and a process for producing efficiently an addition polymer.

[Solution means]

A catalyst for addition polymerization obtained by contacting (A), (B) and (C) described below, and a process for producing an addition polymer with the catalyst:

10 (A) an organoaluminum compound,

(B) one or more of boron compounds selected from (C1) to (C3) below;

(B1) a boron compound represented by the general formula $BQ^1Q^2Q^3$,

15 (B2) a boron compound represented by the general formula $G^+(BQ^1Q^2Q^3Q^4)^-$, and

(B3) a boron compound represented by the general formula $(L-H)^+(BQ^1Q^2Q^3Q^4)^-$.

(wherein B is a boron atom, Q^1 to Q^4 are a halogen atom, a hydrocarbon group, a halogenated hydrocarbon group, a heterocyclic group, a substituted silyl group, an alkoxy group, or a di-substituted amino group, and they may be the same or different; G^+ is an inorganic, organic or organometallic cation; L is a neutral Lewis base, and $(L-H)^+$ is a Brønsted acid.), and

25

(C) a solid inorganic compound.

[Selected Drawing] None

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